

Magnetic relaxation in uranium ferromagnetic superconductors

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There is developed a phenomenological theory of quasi-elastic neutron scattering in the itinerant ferromagnets UGe₂ and UCoGe based on their property that magnetization supported by the moments located at uranium atoms is not conserved quantity relaxing to equilibrium by the interaction with itinerant electrons subsystem. As result the line width of quasi elastic neutron scattering at $q \rightarrow 0$ acquires nonvanishing value at all temperatures but the Curie temperature.

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The family of heavy fermionic uranium metallic compounds UGe₂, URhGe and UCoGe possesses astonishing property of coexistence of superconductivity and ferromagnetism (for the most recent reviews see^{1,2}). Ferromagnetism does not suppress the superconductivity with triplet pairing and since the discoveries of superconductivity in uranium ferromagnets they were considered as equal spin pairing superconductors similar to ³He-A and ³He-A₁ superfluids. The pairing interaction in liquid helium is due to spin-fluctuation exchange, hence, it was quite natural to consider the same mechanism as the origin of superconductivity in uranium compounds. There was implied that they are fully itinerant ferromagnets and the same 5*f* electrons are responsible for ferromagnetism and superconductivity. Thus, practically in all publications the uranium ferromagnet superconductors were considered in frame of theory of the isotropic Fermi liquid with ferromagnetism induced by the Landau-Stoner interaction between electrons (one can find the list of corresponding references in review¹). This beautiful theoretical model is reasonable for ³He. But in relation to the uranium compounds its applicability is quite doubtful in view of significant crystallographic and magnetic anisotropy, as well because of non-itinerant nature of magnetism in these materials. Leaving for following publications the superconducting properties we discuss here the long standing problem of magnetic excitations³ which was one of many unresolved puzzles created in attempt to interpret observations in frame of the Fermi liquid approach.

The magnetic excitations reveal themselves in neutron scattering measurements of dynamical structure factor

$$S_{\alpha\beta}(\mathbf{q}, \omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} \langle M_{\alpha\mathbf{q}}(t) M_{\beta-\mathbf{q}}(0) \rangle_{eq}$$

which is wave vector - frequency dependent magnetic moments correlation function⁴. For an isotropic ferromagnet $S_{\alpha\beta}(\mathbf{q}, \omega) = S(\mathbf{q}, \omega) \delta_{\alpha\beta}$. In absence of walls and spin-orbital coupling the magnetization is conserved quantity, hence, in Heisenberg ferromagnet above Curie temperature the only mechanism leading to the magnetization relaxation is the spin diffusion that results in^{4,5}

$$S(\mathbf{q}, \omega) = \frac{2\omega\chi(\mathbf{q})}{1 - \exp(-\frac{\omega}{T})} \frac{Dq^2}{\omega^2 + (\Gamma_{\mathbf{q}}^d)^2}, \quad (1)$$

such that line width of quasi elastic scattering

$$\Gamma_{\mathbf{q}}^d = Dq^2 \quad (2)$$

is determined by the diffusion coefficient D . We put the Planck constant $\hbar = 1$. The q^2 law dependence was observed in wide temperature range above T_c in Ni and Fe (see⁶ and references therein) reducing at $T = T_c$ to $\Gamma \propto q^{2.5}$ dependence according to predictions of mode-mode coupling theory⁷.

In weak itinerant ferromagnets above Curie temperature another mechanism of dissipationless relaxation can dominate with structure factor given by^{8,9}

$$S(\mathbf{q}, \omega) = \frac{2\omega\chi(\mathbf{q})}{1 - \exp(-\frac{\omega}{T})} \frac{\Gamma_{\mathbf{q}}^L}{\omega^2 + (\Gamma_{\mathbf{q}}^L)^2}, \quad (3)$$

here $\chi(\mathbf{q}) = \frac{\chi_0 k_F^2}{\xi^{-2} + q^2}$, χ_0 is the Pauli susceptibility and the line width $\Gamma_{\mathbf{q}}^L$ is determined by equality

$$\chi(\mathbf{q})\Gamma_{\mathbf{q}}^L = \chi_0\omega(\mathbf{q}) \quad (4)$$

where $\omega(\mathbf{q})$ is the Landau damping frequency equal to $\frac{2}{\pi}qv_F$ for the spherical Fermi surface. The linear in wave vector line width was observed in MnSi¹⁰, however, in the other weak itinerant ferromagnets MnP¹¹ and Ni₃Al¹² the line width q -dependence is closer to the dynamic scaling theory predictions⁷.

The investigations of magnetic excitations in UGe₂ and UCoGe has been reported in several publications^{3,13,14}. The main result is that $\Gamma_{\mathbf{q}}$ unlike both Eqs (2) and (4) does not vanish as $q \rightarrow 0$ for temperatures different from T_c .¹⁵ Here we propose the phenomenological explanation of this observation based on specific nature of 5*f* electron itineracy and localization in ferromagnet and paramagnet states. To be more concrete we shall discuss mostly UGe₂.

Magnetic susceptibility of single UGe₂ crystals has been measured by Menovsky et al¹⁶ (for the more recent results see papers^{17,18}). The easy axis magnetization at zero temperature was found 1.43 μ_B /f.u. that in case of itinerant ferromagnetism corresponds to completely polarized single electron band. On the other hand the neutron scattering measurements of magnetic form factor¹⁹ shows that: (i) the shape of its q dependence is

not distinguishable from the wave vector dependences of the form factors of free U^{3+} or U^{4+} ions, (ii) practically whole magnetic moment both in paramagnetic and ferromagnetic states concentrated at uranium atoms²⁰ and (iii) its low temperature value at $q \rightarrow 0$ coincides with magnetization measured by magnetometer with accuracy of the order 1 percent. These observations unequivocally point on the local nature of UGe_2 ferromagnetism.

The configuration of localized $5f^n$ electrons of each atom of UGe_2 in paramagnetic state mostly consists of superposition of three quasidoublets and three singlets arising from the state with fixed value of total momentum $J = 4$ split by the crystal field.¹⁸ The temperature decrease causes the change in probabilities of populations of crystal field states revealing itself in temperature dependence of magnetic moment. The quasidegenerate ground state formed by the lower quasidoublet allows the system to order magnetically with the ordered moment of $\sim 1.5\mu_B$ twicely smaller than Curie-Weiss moment deduced from susceptibility above the Curie temperature²¹.

The itinerant electron subsystem formed by $7s$, $6d$ and partly $5f$ electrons is also present providing about $0.02\mu_B$ long range magnetic correlations as demonstrated by muon spin relaxation measurements^{22,23}. The interaction between localized and itinerant electron subsystems leads to the magnetization relaxation measured by neutron scattering in paramagnetic and ferromagnetic state. This type of relaxation can be considered as analog of spin-lattice relaxation well known in physics of nuclear magnetic resonance²⁴. In our case the magnetization created by the local moments of uranium atoms plays the role of "spin" subsystem, whereas the itinerant electrons present the "lattice" degrees of freedom absorbing and dissolving fluctuations of magnetization. According to this, we shall treat the total magnetization almost completely determined by the local moments of uranium atoms as not conserved quantity. A deviation of magnetization from the equilibrium value can be transferred to the itinerant electrons and then passed away by spin diffusion which is assumed as much faster in the itinerant than in the localized moment subsystem.

Let us discuss first relaxation above the Curie temperature. The relaxation rate of the order parameter fluctuation is determined by deviation of system free energy

$$\mathcal{F} = \int dV \left(F_h + K_{ij} \frac{\partial M_\alpha}{\partial x_i} \frac{\partial M_\alpha}{\partial x_j} \right) \quad (5)$$

from equilibrium. UGe_2 crystallizes in the orthorhombic structure. The magnetic ordering occurs along a crystallographic direction, and the homogeneous part of the free energy density is

$$F_h = \alpha_x(T)M_x^2 + \alpha_y M_y^2 + \alpha_z M_z^2, \quad (6)$$

$$\alpha_x(T) = \alpha_{x0} \frac{T - T_c}{T_c}, \quad (7)$$

$\alpha_y > 0$, $\alpha_z > 0$, whereas gradient energy in orthorhombic crystal written in exchange approximation²⁵ is determined by three nonzero constants K_{xx}, K_{yy}, K_{zz} . The coordinates x, y, z correspond to the a, b, c crystallographic directions.

To describe new type of relaxation together with diffusion we shall use set of kinetic equations²⁶ relating to each magnetization component

$$\frac{\partial M_\alpha}{\partial t} = -A_{\alpha\beta} \frac{\delta \mathcal{F}}{\delta M_\beta}, \quad (8)$$

where the kinetic coefficient matrix has three nonzero elements A_{xx}, A_{yy}, A_{zz} . One can rewrite the above equations as

$$\frac{\partial M_\alpha}{\partial t} + \nabla_i j_{\alpha i} = -\frac{M_\alpha}{\tau_\alpha}, \quad (9)$$

where $\tau_x^{-1} = 2A_{xx}\alpha_{x0}\frac{T-T_c}{T_c}$, $\tau_y^{-1} = 2A_{yy}\alpha_y$, $\tau_z^{-1} = 2A_{zz}\alpha_z$ and there is no summation over the repeating indices in the right hand side of this equation. The components of spin diffusion currents are

$$j_{\alpha i} = -2A_{\alpha\beta}K_{ij}\frac{\partial M_\beta}{\partial x_j}. \quad (10)$$

Measurements reported in the paper³ with scattering wave vector \mathbf{q} parallel to the crystal a axis revealed no extra scattering relative to the background while for the \mathbf{q} parallel to the c -axis ($\mathbf{q} \parallel \hat{z}$) a strongly temperature dependent contribution was found. The treatment similar to that was used to get the diffusion scattering function⁵ given by Eqs.(1),(2) yields

$$S_{xx}(q_z, \omega) = \frac{2\omega\chi_{xx}(q_z)}{1 - \exp(-\frac{\omega}{T})} \frac{\Gamma_{q_z x}}{\omega^2 + \Gamma_{q_z x}^2}, \quad (11)$$

and the same structure expressions for $S_{yy}(q_z, \omega)$ and $S_{zz}(q_z, \omega)$ correlators. The corresponding widths of quasi-elastic scattering are

$$\Gamma_{q_z x} = 2A_{xx}(\alpha_x(T) + K_{zz}q_z^2), \quad (12)$$

$\Gamma_{q_z y} = 2A_{yy}(\alpha_y + K_{zz}q_z^2)$, and $\Gamma_{q_z z} = 2A_{zz}(\alpha_z + K_{zz}q_z^2)$. The correlator $S_{xx}(q_z, \omega)$ having a form characteristic of critical magnetic scattering contributes the main part in differential cross section of scattering. As one can see $\Gamma_{q_z x}$ does not vanish as $q_z \rightarrow 0$ for temperatures different from T_c in correspondence with the results reported in the paper³. This property is the consequence of the relaxation mechanism specific for heavy fermionic ferromagnet uranium compounds where magnetization created by the moments located at uranium atoms relaxing to equilibrium by the interaction with itinerant electrons subsystem.

Below Curie temperature in the ferromagnetic state the deviation magnetization from equilibrium value $M = M(T)$ is $(M_x - M, M_y, M_z)$. The homogeneous part of free energy density of magnetic fluctuation is

$$F_h = 2|\alpha_x(T)|(M_x - M)^2 + \alpha_y M_y^2 + \alpha_z M_z^2. \quad (13)$$

One can write kinetic equation similar to (9) only for the magnetization component parallel to ferromagnetic ordering

$$\frac{\partial(M_x - M)}{\partial t} + \nabla_i j_{xi} = -\frac{M_x - M}{\tau_x}, \quad (14)$$

with the same expression for the diffusion current as in paramagnet state. Dynamics of perpendicular to equilibrium magnetization components of magnetization is described by linearized Landau-Lifshitz-Gilbert equations^{25,27,28}

$$\begin{aligned} \frac{1}{\gamma} \frac{\partial(M_y + aM_z)}{\partial t} &= -H_z M_z + h_z(t), \\ \frac{1}{\gamma} \frac{\partial(M_y - aM_z)}{\partial t} &= H_y M_y - h_y(t). \end{aligned} \quad (15)$$

Here γ is gyromagnetic ratio, a is dimensionless damping parameter, $H_y = M(K_{ij}q_i q_j + |\alpha_x| + \alpha_y)$, $H_z = M(K_{ij}q_i q_j + |\alpha_x| + \alpha_z)$ are the components of "effective field"²⁵, $h_y(t)$, $h_z(t)$ are the components of time dependent transverse external field. This set of equations determines the spin-wave spectrum which has particular simple form in the absence of damping $\omega = \gamma \sqrt{H_y H_z}$.

These equations also determine the (\mathbf{q}, ω) dependences of yy and zz components of magnetic susceptibilities. In low frequency limit they are frequency independent and pure real: $\chi_{yy} = H_y^{-1}$, $\chi_{zz} = H_z^{-1}$. The latter means that according to the fluctuation-dissipation theorem they do not make a contribution to the corresponding components of dynamical structure factor determining the cross-section of neutron scattering. Thus, at $T < T_c$ the structure factor is given by the same formula (11) as in paramagnet state, but the width of quasi-elastic scattering now is given by

$$\Gamma_{q_z x} = 2A_{xx} (2|\alpha_x(T)| + K_{zz}q_z^2). \quad (16)$$

The equations (12) and (16) are the main results of the paper. The line width of quasielastic neutron scattering near the Curie temperature proves to be linear function of $T - T_c$. The absolute value of the derivative $|d\Gamma_{q_z x}/dT|$ in ferromagnetic region is roughly twice as large as the corresponding derivative in paramagnetic region. The dependence of the wave vector q_z is parabolic. All of these findings are in qualitative correspondence with the experimental observations reported in the paper³ (see Fig. 4 in this paper).

Recently there was pointed out another possibility possessing the similar property of nonvanishing line width $\Gamma(q \rightarrow 0) \neq 0$ of quasi elastic neutron scattering based on

the dissipationless mechanism of magnetic relaxation in weak itinerant ferromagnets with structure factor given by Eqs. (3), (4).²⁹ It is the Landau damping in ferromagnetic metal with intersection of Fermi surfaces related to different Brillouin bands. Namely, when the Fermi surfaces of two bands intersect each other along a line l the Landau damping $\omega_{12}(\mathbf{q})$ at $q = 0$ acquires finite value³⁰

$$\omega_{12}(\mathbf{q} = 0) \approx N_0 \left(\oint \frac{dl}{(2\pi)^3 |\mathbf{v}_1 \times \mathbf{v}_2|} \right)^{-1} \approx \varepsilon_F. \quad (17)$$

Here vectors \mathbf{v}_1 and \mathbf{v}_2 are the Fermi velocities on the Fermi sheets 1 and 2 at point \mathbf{k}_l at line l . This result is applicable to the pure itinerant ferromagnets with band intersection. The intersection of the different band Fermi surfaces can be established from the ab initio calculations. For uranium compounds they have been already performed³¹⁻³⁴ although without special attention to this problem. The band intersection can be established from the general symmetry considerations in case of metals with nonsymmorphic crystal symmetry containing the symmetry elements including combinations of rotation or reflection operations with translations³⁵⁻³⁷. The situation with uranium ferromagnetic compounds is more complicated because the corresponding symmetry considerations have to take into account the spin-orbital interaction and the absence of time reversal symmetry. Whether or not the band intersection takes place we think that Landau damping mechanism does not determine magnetic relaxation in substances where ferromagnetism arises due to the ordering of magnetic moments located at uranium atoms.

Finally we remark that localized nature of magnetism in orthorhombic uranium ferromagnetic compounds put forward as the most plausible pairing mechanism the interaction between spin waves and conduction electrons. The first such type model has been applied to the superconducting antiferromagnet UPd_2Al_3 ³⁸ and then quite recently to the reentrant ferromagnetic superconductor URhGe ³⁹.

In conclusion we developed a phenomenological description of quasi-elastic magnetic relaxation based on specific for heavy fermionic ferromagnet uranium compounds property that magnetization supported by the moments located at uranium atoms is not conserved quantity relaxing to equilibrium by the interaction with itinerant electrons subsystem. As result the line width of quasi elastic neutron scattering at $q \rightarrow 0$ acquires nonvanishing value at all temperatures besides the Curie temperature.

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